

Giant non-linearities accompanying electromagnetically induced transparency

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We develop a fully quantum treatment of electromagnetically induced transparency (EIT) in a vapor of three-level Λ -type atoms. Both the probe and coupling lasers with arbitrary intensities are quantized, and treated on the same footing. In addition to reproducing known results on ultraslow pulse propagation at the lowest order in the ratio of their Rabi frequencies, our treatment uncovers that the atomic medium with EIT exhibits giant Kerr as well as higher order non-linearities. Enhancement of many orders of magnitude is predicted for higher-order refractive-index coefficients. PACS numbers: 42.50.Gy, 42.65.-k, 42.65.An

Introduction The discovery of electromagnetically induced transparency (EIT) [1] has led to the observation of new effects and development of new techniques in quantum optics. Recent examples include ultraslow light pulse propagation [2,3] and light storage [4,5] in atomic vapor, and atomic ground state cooling [6]. It was proposed [7] that enhancement of efficiencies in non-linear optical processes may be achieved by using EIT. A giant cross-Kerr non-linearity in EIT was suggested by Schmidt and Imamoglu [8], and has been indirectly measured in the experiment [2]. We also note representative publications [9] utilizing EIT to study various optical non-linearities.

Essential to all these techniques is the phenomenon of EIT, in which coherent population trapping [10] is induced by atomic coherence and interference. As is well-known, a vapor of three-level Λ -type atoms, initially in the ground state, when irradiated by two laser beams (the coupling and probe laser), exhibits EIT under certain conditions. Conventionally, both the coupling and probe lasers were treated as classical, external fields. In such treatments, the occurrence of EIT requires the coupling laser be much stronger than the probe laser. However, in the experiment [2] this condition was only marginally satisfied, which did not seem to have affected EIT. Moreover, with a quantum treatment of the probe laser, Fleischhauer and Lukin [11] have recently been able to predict the possibility of coherently controlling the propagation of light pulses via dark-state polaritons, which are formed through quantum entanglement of atomic and probe-photon states. This possibility is realized in the latest light storage experiments [4,5]. This lesson teaches us that the quantum description of laser is more fundamental than the classical one, having advantages in uncovering new effects involving quantum nature of photons.

As a natural next step in this direction, in the present Letter we initiate a fully quantum treatment of EIT, namely we will deal with two quantized (coupling and probe) photon field modes interacting with three-level Λ -type atoms. In this treatment the probe and coupling lasers are more or less on the same footing. This will have advantages in studying non-linearities in EIT, which requires the capability to deal with higher orders in the ratio of probe to coupling laser strengths (or the ratio of

Rabi frequencies). Indeed, we will see that in addition to reproducing the well-known results on ultraslow light pulse propagation in atomic vapor at the lowest order, our treatment uncovers giant higher-order optical non-linearities in the atomic medium with EIT. They give rise to dramatic enhancement of Kerr as well as higher-order refractive-index coefficients.

The Model and Dressed States Let us consider a three-level atom, with energy levels $E_1 < E_3 < E_2$, interacting with two quantized laser fields, in the Λ -type configuration (see Fig. 1): The lower two levels $|1\rangle$ and $|3\rangle$ are coupled to the upper level $|2\rangle$. Initially the atom is in the ground state $|1\rangle$. First applied is the coupling laser of frequency $\omega_2 = (E_2 - E_3)/\hbar$. It prepares necessary atomic coherence allowing the later applied probe laser of frequency $\omega_1 = (E_2 - E_1)/\hbar - \Delta_1$ to pass through the atomic medium without much absorption, even if the detuning Δ_1 is zero. This is the EIT [1] underlying the experimentally observed ultraslow pulse propagation in atomic vapor [2,3]. In the following we will present a new treatment of EIT, in which both the probe and the coupling lasers are quantized.

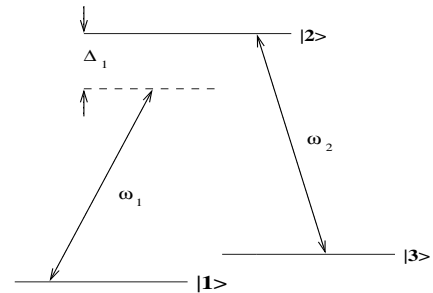


Fig. 1. Energy levels for a Λ -type atom.

With a unitary transformation $U(t) = \exp(-iH_0t/\hbar)$, with $H_0 = \sum_m E_m |m\rangle\langle m| - \hbar[\Delta_1(|2\rangle\langle 2| + |3\rangle\langle 3|) + \omega_1 \hat{a}_1^\dagger \hat{a}_1 + \omega_2 \hat{a}_2^\dagger \hat{a}_2]$, we adopt an interaction picture, in which the Hamiltonian of the atom-field system in the rotating-wave approximation remains time-independent:

$$\hat{H} = \hbar\Delta_1|2\rangle\langle 2| + \hbar\Delta_1|3\rangle\langle 3| + \hbar(g_1\hat{a}_1|2\rangle\langle 1| + g_2\hat{a}_2|2\rangle\langle 3| + H.c.), \quad (1)$$

where $|m\rangle$ ($m = 1, 2, 3$) are atomic states, g 's the coupling constants, and \hat{a}_j and \hat{a}_j^\dagger ($j = 1, 2$) the annihilation and creation operators of the probe and coupling laser modes. Assuming the detuning Δ_1 is small, we can develop a perturbation theory in which we retain only terms linear in Δ_1 . The eigenstates of the Hamiltonian (1), also called the dressed states, have the following form:

$$|\phi_{n_1, n_2}^{(i)}\rangle = a_i|1, n_1, n_2\rangle + b_i|2, n_1 - 1, n_2\rangle + c_i|3, n_1 - 1, n_2 + 1\rangle, \quad (2)$$

where $i = \pm, 0$. The coefficients are given by

$$a_{\pm} = \frac{\Omega_1}{\sqrt{2}\Omega} \left[1 \mp \frac{\Omega_1^2 + 4\Omega_2^2}{2\Omega^3} \Delta_1 \right], \quad (3)$$

$$b_{\pm} = \pm \frac{1}{\sqrt{2}} \left[1 \pm \frac{\Omega_1^2}{2\Omega^3} \Delta_1 \right], \quad c_{\pm} = \frac{\Omega_2}{\sqrt{2}\Omega} \left[1 \pm \frac{3\Omega_1^2}{2\Omega^3} \Delta_1 \right],$$

and

$$a_0 = \frac{\Omega_2}{\Omega}, \quad b_0 = \frac{2\Omega_1\Omega_2}{\Omega^3} \Delta_1, \quad c_0 = -\frac{\Omega_1}{\Omega}, \quad (4)$$

with $\Omega_1 = 2g_1\sqrt{n_1}$, $\Omega_2 = 2g_2\sqrt{n_2+1}$ and $\Omega = \sqrt{\Omega_1^2 + \Omega_2^2}$. The energy eigenvalues of the dressed states (2) are

$$E_{n_1, n_2}^{(\pm)} = \frac{\Omega_1^2 + 2\Omega_2^2}{2\Omega^2} \Delta_1 \pm \frac{\Omega}{2}, \quad E_{n_1, n_2}^{(0)} = \frac{\Omega_1^2}{\Omega^2} \Delta_1. \quad (5)$$

The complete set of dressed states for the system under consideration comprises the states $|\phi_{n_1, n_2}^{(i)}\rangle$ ($i = \pm, 0$) for each $n_1 > 0$, and $n_2 \geq 0$ together with other two states $|1, 0, n_2\rangle$ and $|3, n_1, 0\rangle$ with zero eigenvalues. Knowing this set allows us to determine the time evolution of the atom-field system for any initial configuration.

Steady State and Susceptibility Assume that the atom is initially in the ground state, while the coupling and probe lasers in a coherent state $|\alpha, \beta\rangle$, with α and β supposed to be real for simplicity. Namely the initial state of the atom-field system is assumed to be

$$|\Psi(0)\rangle = |1\rangle \otimes |\alpha, \beta\rangle. \quad (6)$$

In the usual treatment with both lasers being classical external fields, one is concerned with a steady state of the atomic system. The counterpart of the steady state in our fully quantum treatment, according to the commonly used *adiabatic hypothesis* in quantum scattering theory and quantum field theory [12], is the state that evolves from the initial state (6) with the couplings g_1 and g_2 *adiabatically turned on*. Physically this is equivalent to having localized laser pulses before they enter the atomic vapor, with the pulse shape sufficiently smooth. In conformity to the Adiabatic Theorem, we need to identify a linear combination of the dressed states (2) that tends to the initial state (6) if we take the limits $g_1, g_2 \rightarrow 0$ (or $\Omega_1, \Omega_2 \rightarrow 0$). According to Eqs. (3-5), the ordering of the

limits $\Omega_1 \rightarrow 0$ and $\Omega_2 \rightarrow 0$ is important. Corresponding to the actual conditions in which EIT is observed, the correct ordering is first $\Omega_1 \rightarrow 0$ and then $\Omega_2 \rightarrow 0$. In our interaction picture, this procedure selects the $i = 0$ state in Eq. (2). Transforming it back to the Schrödinger picture, we identify the following state as the state that evolves adiabatically from the initial state:

$$|\Psi(t)\rangle = \sum_{n_1, n_2=0}^{\infty} e^{-(\alpha^2+\beta^2)/2} e^{-iEt} \frac{\alpha^{n_1} \beta^{n_2}}{\sqrt{n_1! n_2!}} |\phi_{n_1, n_2}^{(0)}\rangle, \quad (7)$$

with $E = E_1 + \omega_1 n_1 + \omega_2 n_2 + E_{n_1, n_2}^{(0)}$. This state can be viewed as the counterpart of the usual "steady" state in our treatment. We see that the population of the upper level $|2\rangle$ is zero up to first order in detuning Δ_1 . This means that there is no absorption, implying the phenomenon of EIT. We note that for EIT to occur the usual treatment requires the coupling laser be much stronger than the probe one [13], i.e. $\Omega_2 \gg \Omega_1$; then most atoms are populated in the ground state. Our formalism does not require this for EIT, so broader conditions are allowed: The coupling and probe lasers can be equally strong or even the probe laser is stronger. Then atoms can be in a more general superposition of two atomic states, which are entangled with the coupling and probe fields.

From Eq. (7) one can form the total density operator of the atom-field system. After taking trace over the field states, one gets the atomic reduced density operator. Then the Fourier component of the optical coherence of the "steady" state at the frequency of the probe laser is found to be

$$\rho_{21}^A(\omega_1) = a_0(\bar{n}_\alpha, \bar{n}_\beta) b_0(\bar{n}_\alpha, \bar{n}_\beta), \quad (8)$$

where $\bar{n}_\alpha = \alpha^2$, and $\bar{n}_\beta = \beta^2$ are, respectively, the mean photon numbers of the coupling and probe lasers in the coherent state $|\alpha, \beta\rangle$. In the derivation of Eq. (8), we have used the large- n approximation, i.e., $\bar{n}_\alpha, \bar{n}_\beta \gg 1$, so that the photon distributions are sharply peaked around their mean value. (For example, in a recent experiment of light storage [4], $\bar{n} \sim 10^4$.)

The polarization of the atomic medium at the frequency of the probe laser is determined to be

$$P(\omega_1) = \mu_{21} N \rho_{21}^A(\omega_1) = \epsilon_0 \chi(\omega_1) E_1(\omega_1), \quad (9)$$

where N is the number density of atoms, μ_{21} the transition dipole moment between states $|2\rangle$ and $|1\rangle$, ϵ_0 the free space permittivity, $\chi(\omega_1)$ the susceptibility of the atomic medium, $E_1(\omega_1)$ the Fourier component of the mean electric field for the probe laser at frequency ω_1 . The value of $E_1(\omega_1)$ can be calculated from the "steady" state (7). In the large- n approximation, $E_1(\omega_1) = \mathcal{E}_1 \alpha$, where $\mathcal{E}_i = (\hbar \omega_i / 2 \epsilon_0 V)^{1/2}$ ($i = 1, 2$) with V the quantized volume. Then the susceptibility is given by

$$\chi(\omega_1) = \frac{4N|\mu_{12}|^2\bar{\Omega}_2^2\Delta_1}{\hbar\epsilon_0(\bar{\Omega}_1^2 + \bar{\Omega}_2^2)^2}, \quad (10)$$

where $\bar{\Omega}_1 = \Omega_1(\bar{n}_\alpha, \bar{n}_\beta)$ and $\bar{\Omega}_2 = \Omega_2(\bar{n}_\alpha, \bar{n}_\beta)$ are the Rabi frequencies of the coupling and probe lasers, respectively. This is the main result of this paper, valid for arbitrary ratio of $\bar{\Omega}_1/\bar{\Omega}_2$.

Eq. (10) exhibits the signature of EIT: the linear susceptibility vanishes at the resonance ($\Delta_1 = 0$). The derivative of $\chi(\omega_1)$ is related to the group velocity for the probe laser pulse: $v_g = c/[1 + (\omega_1/2)(d\chi/d\omega_1)]$, with c speed of light in vacuum. Thus we have

$$v_g = v_g^0 \frac{(\bar{\Omega}_1^2 + \bar{\Omega}_2^2)^2}{\bar{\Omega}_2^4}, \quad (11)$$

where $v_g^0 = \hbar c \epsilon_0 \bar{\Omega}_2^2 / (2\omega_1 |\mu_{12}|^2 N)$ is the usual expression for the group velocity [2]. This equation shows that in general the group velocity of the probe laser depends on the Rabi frequency of the coupling as well as the probe laser. The $\bar{\Omega}_1$ dependence of the group velocity is a new result of our treatment, closely related to the higher order nonlinear susceptibilities we will discuss below.

To relate the present results to earlier studies of EIT-based dispersive properties [14], we expand the susceptibility given in Eq. (10) in terms of powers of $\bar{\Omega}_1/\bar{\Omega}_2$. When $\bar{\Omega}_1 \ll \bar{\Omega}_2$, the first term of this expansion gives us the linear susceptibility

$$\chi^{(1)}(\omega_1) = \frac{4|\mu_{12}|^2 N \Delta_1}{\hbar \epsilon_0 \bar{\Omega}_2^2}. \quad (12)$$

Thus, at the lowest order we recover results in Ref. [14], when the decay rates of states are neglected at the resonant frequency of the probe laser. Similarly, keeping only the lowest-order term in $\bar{\Omega}_1/\bar{\Omega}_2$, Eq. (11) will give the usual expression, v_g^0 , for the group velocity [2], which does not depend on $\bar{\Omega}_1$ and involves the contributions only from linear susceptibility.

The refractive index of the medium can be obtained from Eq. (10) by definition $n \equiv \sqrt{1 + \chi}$. Making use of Eqs. (10) and (11), we find the refractive index change near zero probe detuning to be

$$\Delta n = \frac{\lambda_1}{2\pi} \frac{\Delta_1}{v_g}, \quad (13)$$

where λ_1 is the wavelength of probe laser. It is worthwhile to note that this refractive index change involves the contributions of all orders of nonlinear susceptibilities due to the intensity dependence of the group velocity in (11). For the slow light experiment [2], with $v_g = 17$ m/s and parameters $\Delta_1 = 1.3 \times 10^6$ rad/s and $\lambda_1 = 589$ nm, we obtain from Eq.(13) $\Delta n = 7.2 \times 10^{-3}$. This value agrees with the measured value in Ref. [2].

Giant Non-linearities Nonlinearities play an important role not only in nonlinear optics but also in quan-

tum optics. They may be used for generation of enhanced squeezing [15], quantum computation and quantum teleportation [16], and quantum nondemolition measurements [17]. Since EIT takes place in the vicinity of atomic resonance, large nonlinearities are naturally expected. We note that in the conventional steady-state approach, it is difficult to obtain nonlinear susceptibilities higher than $\chi^{(1)}$ and $\chi^{(3)}$. However, in our formalism we can get arbitrary higher-order nonlinear susceptibilities once for all.

The higher-order nonlinear susceptibilities are defined by

$$\chi(\omega_1) = \chi^{(1)}(\omega_1) + \chi^{(3)}(\omega_1)|E(\omega_1)|^2 + \chi^{(5)}(\omega_1)|E(\omega_1)|^4 + \chi^{(7)}(\omega_1)|E(\omega_1)|^6 + \dots \quad (14)$$

where $\chi^{(1)}(\omega_1)$ is the linear susceptibility given in Eq. (12), and $\chi^{(k)}(\omega_1)$ ($k \geq 3$) represent the k th-order nonlinear susceptibility. Assuming that the coupling laser is stronger than the probe, we expand the susceptibility (10) in powers of $\bar{\Omega}_1/\bar{\Omega}_2$. Rewriting this expansion in the form of Eq. (14), we find

$$\begin{aligned} \chi^{(3)}(\omega_1) &= -\frac{4\epsilon_0 c}{I_2} \chi^{(1)}(\omega_1), \quad \chi^{(5)}(\omega_1) = -\frac{3\epsilon_0 c}{I_2} \chi^{(3)}(\omega_1), \\ \chi^{(7)}(\omega_1) &= -\frac{8\epsilon_0 c}{3I_2} \chi^{(5)}(\omega_1), \end{aligned} \quad (15)$$

where $I_2 = 2\epsilon_0 \mathcal{E}_2 c \beta^2$ is the intensity of the incident coupling laser.

Normally nonlinear optical properties of the medium are described by the nonlinear refractive indices [18]:

$$n = n_0 + n_2|E_1|^2 + n_4|E_1|^4 + n_6|E_1|^6 + \dots \quad (16)$$

where the first nonlinear correction to the refractive index is the Kerr coefficient n_2 , which is related to $\chi^{(3)}(\omega_1)$. n_k ($k \geq 4$) are higher-order nonlinear refractive-index coefficients, related to higher-order nonlinear susceptibilities up to $\chi^{(k+1)}(\omega_1)$.

Making use of Eqs. (14) and (15), we obtain from Eq. (16) nonlinear refractive index coefficients:

$$\begin{aligned} n_2 &= -\frac{2\epsilon_0 c}{I_2} \chi^{(1)}(\omega_1) = -\frac{2\epsilon_0 c \Delta_1}{\pi I_2} \frac{\lambda_1}{v_g^0}, \\ n_4 &= -\frac{3\epsilon_0 c n_2}{I_2}, \quad n_6 = -\frac{8\epsilon_0 c n_4}{3I_2}. \end{aligned} \quad (17)$$

To increase the value of nonlinear refractive-index coefficients one may either increase the atomic density or decrease the coupling laser intensity.

To demonstrate the magnitude of the giant nonlinearities derived above, we calculate the nonlinear refractive-index coefficients using the parameters ($I_2 = 40$ mW/cm², $\Delta_1 = 1.3 \times 10^6$ rad/s, and $\lambda_1 = 589$ nm) in the ultraslow light experiment reported in Ref. [2], in which a light pulse speed 17 m/s was observed. We estimate that under these conditions, $n_2 = -1.9 \times 10^{-7}$

m^2/V^2 , $n_4 = 3.8 \times 10^{-12} \text{ m}^4/\text{V}^4$, and $n_6 = -6.7 \times 10^{-17} \text{ m}^6/\text{V}^6$. In terms of a common practical unit, $n_2 = -0.36 \text{ cm}^2/\text{W}$, $n_4 = 13.0 \text{ cm}^4/\text{W}^2$, and $n_6 = 4.5 \times 10^2 \text{ cm}^6/\text{W}^3$. The value of the Kerr nonlinearity is of the same order of magnitude as that indirectly measured in Ref. [2], almost 10^6 times greater than that measured in cold Cs atoms [2,19], and $\sim 10^{12}$ times greater than that measured in other materials [20]. The fourth-order refractive-index coefficient n_4 is $\sim 10^{22}$ times greater than that measured in other materials [20].

In optical fibers, the ratio between the second- and the fourth-order refractive-index coefficients is an essential parameter [21] to obtain stable spatial solitary waves. The lower the ratio n_2/n_4 , the lower the required power for stable beam propagation. For the atomic medium with EIT, we have $n_2/n_4 = -I_2/(3\epsilon_0 c)$. With the parameters in the experiment [2], we estimate $|n_2/n_4| \sim 10^{-2} \text{ W/cm}^2$. This ratio is small compared to most other nonlinear media [20] by almost 11 orders of magnitude. From Eq. (17) we also obtain the ratio between the fourth- and the sixth-order refractive-index coefficients $n_4/n_6 = -3I_2/8(\epsilon_0 c)$, which is of the same order as n_2/n_4 .

We note that when the ratio of $\bar{\Omega}_1/\bar{\Omega}_2$ is close to unity, one had better directly deal with the original formula (10), rather than using its power series expansion.

Conclusions We have developed a fully quantum treatment of EIT in atomic medium. Both the probe and coupling lasers are quantized and treated on the same footing. This allows us to deal with the cases in which the ratio of the probe-to-coupling Rabi frequencies is not necessarily small. At the lowest order in this ratio, we are able to reproduce the known results for slow pulse propagation. At higher orders we have uncovered that accompanying the EIT phenomenon, atomic medium possesses giant optical non-linearities, which can give rise to dramatic enhancement of the Kerr as well as higher order refractive-index coefficients. In other words, the atomic medium with EIT is really a very unusual optical medium; to the list of its astonishing optical properties, we now add another remarkable one: giant optical non-linearities. It would be very interesting to observe these giant non-linearities and explore their potential applications.

Contrary to the usual treatment of EIT, our present treatment is essentially a time-independent approach, because both probe and coupling lasers are included as part of the dynamical system. On the other hand, an important limitation is that we have ignored the decay rates of various levels. How to incorporate them in a fully quantum treatment is a challenging issue.

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